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Spin-Lattice Interaction in the Quasi-One-Dimensional Helimagnet $LiCu_2O_2$

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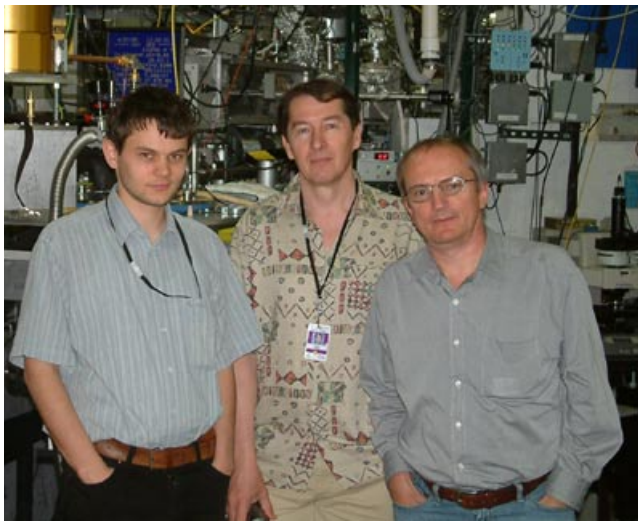
We studied electron spin resonance (ESR) in $LiCu_2O_2$, using the far-infrared/high magnetic field instrumentation at U12IR. The electron spins in this quasi one-dimensional material develop a helical order at low temperature. The excitation of this spin chain results in a field and frequency dependent absorption feature in the IR spectrum. We compared the results to a theoretical analysis of the excitation, and we deduced the characteristic anisotropy energy of the spin system. In particular, we determined the interactions that keep the spins confined to a particular plane relative to the crystal.

Ferromagnets are all around us – in the seal on the door of the refrigerator, in the headset of the iPod, and in the computer's hard disk, to name a few examples. In ferromagnets, the interaction between the electrons makes the electron spins (and the associated magnetic moments) line up parallel to each other. What happens if the interaction is different, favoring spins in opposite directions? These materials, the so-called *antiferromagnets*, do not have widespread applications, but they are much less understood and the subject of intensive current research.

In this study we looked at a quasi one-dimensional antiferromagnet, $LiCu_2O_2$. The copper in this compound has two different ionization states. The Cu^{3+} ions are magnetically neutral, but the Cu^{2+} ions carry magnetic moments with spin ½. These ions form quasi one-dimensional chains with a zigzag "ladders" along the b direction in the crystal (**Figure 1**).

In addition to its quasi one-dimensional structure, this material has two other interesting characteristics: The interaction between the moments goes beyond the first neighbors, and the structure has a triangular motif. In antiferromagnets both of these properties lead to *frustration*: it is impossible to find an arrangement of spins so that all interactions lead to a minimum of the energy. (For example, three spins on a triangle cannot all point opposite to each other.) Because of the absence of a simple classical ground state, quantum effects and fluctuations are expected to be important in this material. However, experiments, primarily NMR and neutron scattering, led to a somewhat disappointing conclusion. Instead of a fancy quantum state, the spins exhibit a helical order along the chains, with a wavevector Q incommensurate to the lattice. In zero external field all spins were found to be parallel to the ***a-b*** plane. (**Figure 2**) The intricate spin order was interpreted in terms of a classical interacting spin model, with no (or very little) quantum fluctuations. Why does this system of spin ½ electrons behave as if quantum mechanics did not exist? This was the principal question in our study.



Bálint Náfrádi, László Forró and László Mihály at U12IR

We searched for the answer by trying to measure the energy gap in the excitation (magnon) spectrum, since the absence of low-energy excitations stabilizes the classical ground state. The main experimental results of the study are shown in the upper panel of **Figure 3**. There is a weak, but visible resonance at finite

frequencies in zero external field. This is a direct measurement of the energy gap, responsible for suppressing quantum fluctuations.

What is the source of the energy gap? And here is a related question: What makes the spins prefer the a-b plane, instead of making a helical structure in some different planes? The anisotropy in interaction between the spins is needed to answer both of these questions. In this case the anisotropy is of the "easy plane" type: The interaction energies are larger for spins in the **a-b** plane than for spins in other directions. We performed theoretical modeling and, by fitting the resonance positions for all field, we determined a single anisotropy parameter for this system. The model also described the observed strong field dependence in the intensity of the resonance.

Interestingly, the model predicted two resonances at finite fields, but the other one (green line in **Figure 3**) was much weaker. We suspected that the instrument we used was not sensitive enough to see this other line, and we sent the sample back to EPFL, (Lausanne, Switzerland), where Bálint Náfrádi did more ESR measurements. The spectrometer in Lausanne operates at a few fixed frequencies, and it cannot be used to create a full two-dimensional map of absorption intensity seen in **Figure 3**, but it is much more sensitive than ours. The resonance line has been found at the predicted position, and we witnessed one of the rare cases when the theory not only interpreted the existing experimental data, but actually predicted the result of a new experiment.

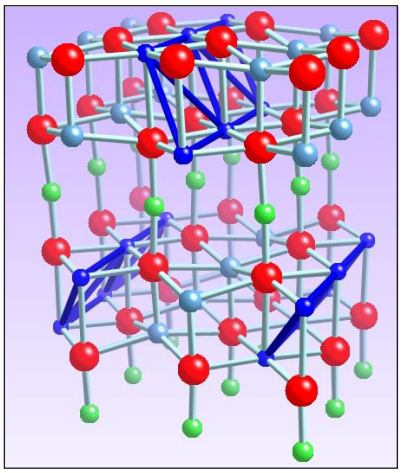


Figure 1. *Crystal structure of $LiCu_2O_2$. The magnetic Cu^{2+} ions are dark blue, the non-magnetic copper is green, the lithium is light blue and the oxygen is red. The dark blue bonds emphasize the triangular spin ladder.*

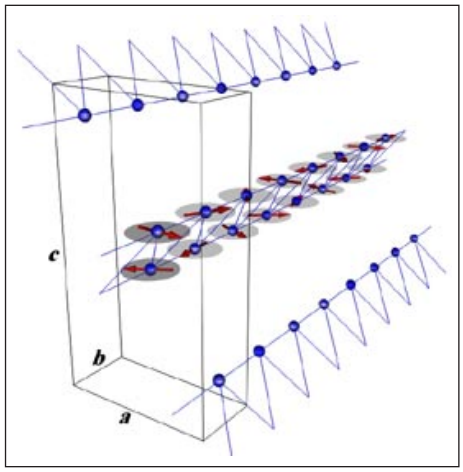


Figure 2. *Helical spin order along a triangular ladder in $LiCu_2O_2$. All spin directions are parallel to the **a-b** plane, and subsequent spins on the same leg of the ladder make an angle slightly larger than 60°.*

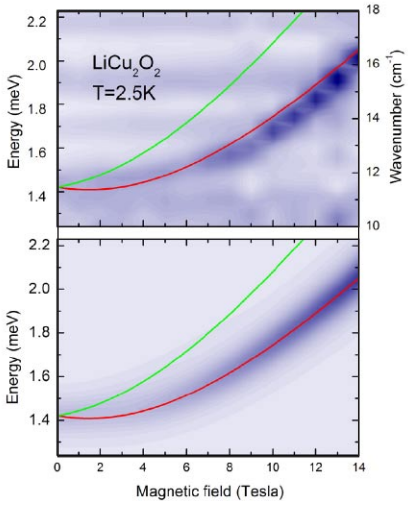


Figure 3. *Upper panel: Absorption due to spin resonance as a function of field and frequency. Darker shades indicate stronger absorption. The red line is a theoretical fit to the line position. The green line is another resonance predicted by the theory. Lower panel: Simulated experimental results, based on the calculated intensity of the absorption.*